# Thermoacoustics for Liquefaction of Natural Gas

By Greg Swift
Los Alamos National Laboratory
and John Wollan
Praxair

A prototype device for liquefying natural gas using thermoacoustics is capable of producing 500 gallons per day of LNG, consuming 35 percent of the incoming gas in the process. Larger capacities operating at higher efficiencies are on the drawing board.

ne ordinarily thinks of a sound wave as consisting only of coupled pressure and position oscillations. In fact, temperature oscillations accompany the pressure oscillations and when there are spatial gradients in the temperature oscillations, oscillating heat flow occurs. The combination of these oscillations produces a rich variety of "thermoacoustic" effects. In everyday life, the thermal effects of sound are too small to be easily noticed; for example, the amplitude of the temperature oscillation in conversational levels of sound is only about 0.0001°C. However, in an extremely intense sound wave in a pressurized gas, these thermoacoustic effects can be harnessed to create powerful heat engines and refrigerators. Whereas typical engines and refrigerators rely on crankshaft-coupled pistons or rotating turbines, thermoacoustic engines and refrigerators have no moving parts (or at most only flexing parts without the need for sliding seals). This simplicity, coupled with reliability and relatively low cost, has highlighted the potential of thermoacoustic devices for practical use. As a result, thermoacoustics is maturing quickly from a topic of basic scientific research through the stages of applied research and on to important practical applications.

In this article, we introduce the basic principles of thermoacoustics and describe progress toward their use for liquefaction of natural gas. Thermoacoustic natural-gas liquefiers are surprisingly simple: They use no exotic materials, require no close tolerances, and are little more than welded pipe and heat exchangers filled with pressurized helium. This simplicity, along with the reliability and low maintenance inherent in thermoacoustic technology, suggests that thermoacoustic liquefiers could enable economic recovery of marginal gas resources such as associated gas from offshore oil wells, gas accumulations at remote locations, and even the recovery of landfill gas and marginal coal seam gas accumulations. In addition, the technology could find an application in areas where smaller-scale gas liquefaction is needed: liquefaction at seasonal peak shaving facilities and at fleetvehicle fueling stations.

#### **Thermoacoustic Basics**

Many varieties of heat-driven thermoacoustic refrigeration systems exist, but in this article we consider only a toroidal thermoacoustic-Stirling hybrid engine driving a thermoacoustic orifice pulse tube refrigerator (Figure 1). Parts (a) and (b) of Figure 1 show

the half-wave resonance present in the apparatus illustrated by the schematic in (c), where the engine is at the top and the refrigerator is at the bottom. Heat exchangers  $(H_v)$  and a regenerator in the engine convert some of the heat power  $(Q_H)$  from burning natural gas at a hot temperature  $(T_H)$  into acoustic power (W), rejecting waste heat power  $(Q_0)$  to a water stream at ambient temperature  $(T_0)$ . Acoustic power is consumed by the refrigerator, which uses it to pump heat  $(Q_c)$  from a liquefying natural-gas load and rejects waste heat  $(Q'_{\theta} + Q''_{\theta})$  to the ambient water stream. Each of the heat exchangers may be of finned-tube or shell-and-tube construction, as open to helium flow as possible. Each regenerator usually consists of a pile of stainless-steel screens, supporting the smooth temperature profile between the two adjacent heat exchangers.

Thermodynamically, acoustic power is just as valuable as other forms of "work" such as electric power or rotating-shaft power. The first law of thermodynamics determines that  $W+Q_0=Q_H$  in the engine. The second law shows that the engine efficiency  $W/Q_H$  is bounded by the Carnot efficiency,  $1-T_0/T_H$ . The most efficient thermoacoustic engine to date has achieved 40 percent of the Carnot efficiency, while the most powerful has

produced 17 kW of acoustic power. Similarly, in the refrigerator, the first law of thermodynamics determines that  $W + Q_C = Q_{\theta}' + Q_{\theta}''$ ; the second law shows that the efficiency  $Q_C/W$ , known as the coefficient of performance, is bounded by the Carnot expression  $T_C/(T_O - T_C)$ . The most efficient thermoacoustic orifice pulse tube refrigerator to date has achieved 25 percent of this Carnot bound.

One of the most important large dimensions in a thermoacoustic device is the length of its resonator, which (together with the helium sound speed) determines the operating frequency, just as the length of an organ pipe determines its pitch. This length typically ranges from 10 cm for the simplest experimental systems to 10 m for today's most efficient and mature systems. The resonator shown in Figure 1

uses a half-wavelength standing wave, shown schematically in parts (a) and (b) (but without details of the wave within the engine and refrigerator). This wave appears spontaneously whenever the temperature in the engine's hot heat exchanger is high enough, and the amplitude of the wave increases as the heat supplied to the hot heat exchanger increases. In parts (a) and (b), the pressure and position waves are shown at two times: the red curves show these variables when the helium is at the uppermost extreme of its position in the resonator, with density and pressure highest at the top of the resonator and lowest at the bottom, while the blue curves show them 180° later in the cycle.

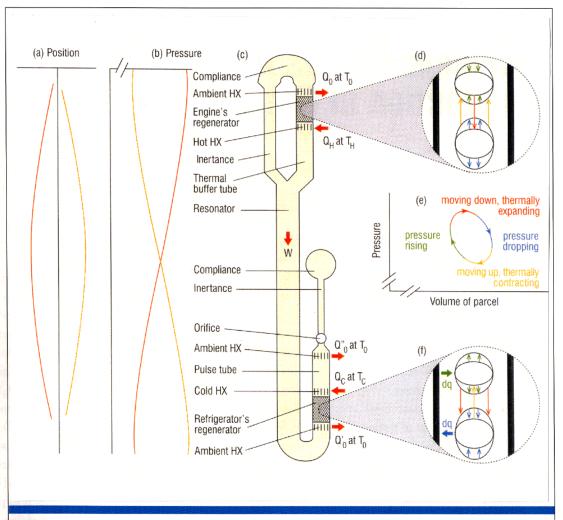


Figure 1: Schematic of a Heat-driven Thermoacoustic Refrigeration System

# **How the Engine Works**

To understand the conversion of heat to acoustic power by this simple engine, consider the magnified view of part of the regenerator shown in Figure 1, part (d), which shows a typical parcel of helium at four instants of time as it oscillates in position, pressure, temperature, and density, exchanging heat with the nearby solid in the regenerator. The tiny pore of the regenerator is shown as a smooth-walled channel for simplicity.

The wave carries the helium up and down along the pore, compressing and expanding it, with time phasing such that it is most pressurized while it is moving down and most depressurized while it is moving up. In typical thermoacoustic engines and refrigerators, the amplitude of the pressure oscillation is 10 percent of the mean pressure, and the amplitude of motion is a similar percentage of the length of the regenerator. Thermal contact between the oscillating helium and the solid wall of the pore, plus the externally imposed temperature gradient, add a new feature to what would otherwise be a simple acoustic oscillation — oscillatory heat transfer between the helium and the solid. While the helium is moving downwards, it encounters ever warmer portions of the regenerator, so it absorbs heat and expands; while the helium is moving upwards, it rejects heat and contracts.

Figure 1, part (e), a pressure-volume

(p-V) diagram for the parcel of helium illustrated in part (d), shows that the helium does net work  $(\int p \ dV)$  on its surroundings because expansion takes during the high-pressure time of the cycle and the contraction during the low-pressure time. This process depends on the correct time phasing between motion and pressure, which is maintained by inertial and compressive effects in the ductwork near the regenerator. The net work that the helium does on its surroundings is produced at the resonance frequency. Thus, the parcel of helium shown in (d), and all others like it within the regenerator, deliver acoustic power to the wave, while the wave sets the frequency of the power production.

Each parcel of helium also deposits a little heat (not shown in Figure 1) at one location in the regenerator while the pressure is rising and the parcel is relatively stationary near the upper extent of its motion. It absorbs that heat near the lower extreme of its motion, at a warmer location in the regenerator, when the pressure is falling. With respect to heat, all parcels act like members of a bucket brigade, with the overall effect being absorption of heat at the hot heat exchanger and rejection of heat at the ambient heat exchanger.

#### **Pore Size**

The pore size in the regenerator determines the nature of the thermal contact between the regenerator solid heat capacity and the moving helium. Good thermal contact is needed to accomplish the cycle shown in Figure 1, because the temperature of the helium should match the local solid temperature while the helium moves. Analysis shows that a spacing between plates of a fraction of a thermal penetration depthd  $\delta_K = \sqrt{K/\pi} f \rho c_p$  is best, where K is the thermal conductivity of the helium,  $\rho$  is its

density,  $c_p$  is its isobaric specific heat per unit mass, and f is the frequency of the acoustic oscillation;  $\delta_K$  is roughly the distance heat can diffuse through the helium during a time  $1/\pi f$ . In today's thermoacoustic systems,  $\delta_K$  is typically a fraction of a millimeter. (Pores too tight impose too much viscous drag on the helium.)

#### **How the Refrigerator Works**

The basic principle of operation of the thermoacoustic orifice pulse tube refrigerator is very similar to that of the thermoacoustic engine. A magnified view of part of the refrigerator's regenerator in Figure 1, part (f), illustrates one typical parcel of helium as it oscillates in position, pressure, temperature, and density, exchanging heat (dq) with the nearby solid in the regenerator, moving that heat up the temperature gradient. As the helium oscillates along the refrigerator's regenerator, it experiences changes in pressure. At the lower extreme of its motion, the typical parcel of helium rejects heat (dq) to the regenerator, because the pressure rises while the helium is relatively stationary at that location. Similarly, at the upper extreme of its motion, it absorbs heat (dq) from the regenerator, because the pressure rises while it is relatively stationary there. Thus, the parcel of helium moves a little heat along the regenerator, up the temperature gradient, during each cycle of the acoustic wave. All the other parcels in the regenerator behave similarly, so that the overall effect, again like in a bucket brigade, is the net transport of heat from the cold heat exchanger to the ambient heat exchanger.

The helium also consumes acoustic power from the wave (not shown in Figure 1), because the thermal expansion of the helium, attending its downward motion, occurs during the

low pressure time of the acoustic wave, and the thermal contraction, attending its upward motion, occurs during the high pressure time. The resulting acoustic power absorbed by the helium is supplied by the thermoacoustic engine, transmitted to the refrigerator through the wave in the resonator.

#### **Development History**

Heat driven acoustic oscillators have been known for over a century — the earliest and simplest was discovered accidentally by European glassblowers. But an accurate theory applicable to thermoacoustic phenomena was not developed until the 1970s, through the efforts of Nicholas Rott and coworkers at ETH-Zurich. Rott's theory is based on a low-amplitude linearization of the Navier-Stokes, continuity, and energy equations, with sinusoidal oscillations of all variables.

In the early 1980s, the thermalphysics team at Los Alamos, supported by BES in DOE's Office of Science, was frustrated by the large number of precision moving parts required for their experiments on the thermodynamic behavior of near-critical liquids in heat engines. While looking for simpler engine designs, they read the publications of Peter Ceperley at George Mason University, who had realized that the timing between pressure changes and motion in Stirling engines is the same as in a traveling sound wave (Ceperley, 1979). Inspired by his insight, the Los Alamos researchers began considering acoustic technology to eliminate moving parts. Eventually, they brought together a thermodynamic point of view, acoustic techniques, explicit heat exchangers, and Rott's theory, producing the first powerful thermoacoustic engines and the first thermoacoustic refrigerators. Fundamental research on

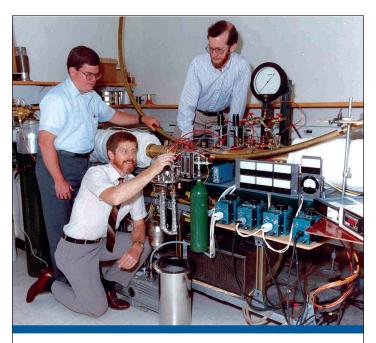


Figure 2: First Thermoacoustically Driven Pulse Tube Refrigerator

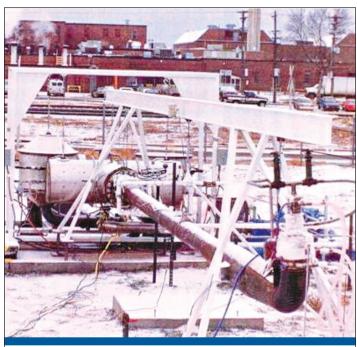
thermoacoustics has grown ever since, at Los Alamos and throughout the world.

In the late 1980s, a partnership between the Los Alamos team and Ray Radebaugh at the National Bureau of Standards (now National Institute of Standards and Technology) in Boulder combined a thermoacoustic engine with an orifice pulse tube refrigerator to create the first cryogenic refrigerator with no moving parts (Figure 2). This device was dubbed the "Coolahoop" because the bent brass portion of the half-wavelength acoustic resonator (extending upward in Figure 2) resembled a hulahoop (Radebaugh, et al., 1991). In the photo, the pulse tube refrigerator is the silver-colored "U" at bottom center, while the two thermoacoustic engines are under the bulky white insulation to the right and left of the refrigerator.

Even though this early system had only 5 W of cooling power at 120 Kelvin, Radebaugh believed from the outset that the best application for this heat-driven refrigerator would be liquefaction of natural gas, using combustion of gas as the heat source. A typical modern gas liquefaction plant costs a billion dollars, liquefies 10<sup>4</sup> m<sup>3</sup>/day, and has substantial operating and maintenance costs. The need for

relatively small, reliable, inexpensive liquefaction equipment seemed clear and an "acoustic liquefier" seemed to fit the need perfectly. The goal of an acoustic liquefier with a capacity of 10,000 gallons per day (gpd) followed, eventually including economic analysis for arrays of such liquefiers on floating LNG production/ storage vessels and oil/gas separation vessels (van Wijngaarden, 1999; Figure 3).

*Cryenco*, a small manufacturing company in Denver, began working on



**Figure 4: First Acoustic Liquefier** 





Figure 3: Illustrations of an Acoustic Liquefier and Offshore Applications

this technology in 1994. The following year, DOE's Office of Fossil Energy (through NETL) began supporting Los Alamos team's partnership with *Cryenco*. Hardware development continued in Denver through many transitions, most recently as Praxair acquired the project. While working on the Denver development, the Los Alamos researchers have continued research on fundamentals, increasing engine efficiency and bringing thermoacoustic improvements to orifice pulse tube refrigerators.

## **Prototype Acoustic Liquefier Hardware**

The first natural-gas-fired thermoacoustic liquefier was completed in Denver in 1997 (Figure 4). It achieved a liquefaction capacity of 140 gpd of LNG, producing 2 kW of refrigeration power at  $-140^{\circ}$ C.

The second phase of hardware development, which began in mid 1999, has been the development of an efficient 500 gpd system (Figure 5). The thermoacoustic portion of the system is prominently visible in Figure 5, with the engine on top and refrigerators on the bottom, linked by a half-wave resonator. The natural gas burner is at the very top, under the blue banner. The engine is in the large bulge below the burner. The refrigerators are hidden inside the large, cylindrical vacuum insulation can near the bottom, but two of their slender inertances and compliances are visible above the vacuum can. The thermoacoustic working helium is at an average pressure of 450 psi, with oscillations up to ±45 psi in amplitude at a frequency of 40 Hz.

In this system, three refrigerators are used, driven in parallel by the thermoacoustic wave but connected in series with respect to the natural-gas stream so that the first acts as a naturalgas precooler, the second removes the rest of the sensible heat and some of the latent heat, and the third removes the rest of the latent heat. The design calls for the engine and resonator to deliver 30 kW of acoustic power to the refrigerators, whose combined cooling power is 7 kW. The burner delivers heat to the engine, and is made more efficient by a traditional recuperator to preheat the incoming fresh air by capturing heat from the flue. Waste heat is removed from the engine and the refrigerators by circulating water at ambient temperature. Overall system efficiency should yield liquefaction of



Figure 5: Current Protoype 500 GPD Acoustic Liquefier

65 percent of a natural-gas stream while burning 35 percent.

In 2001, the 500-gpd system was operated at 60 percent of its design pressure amplitude, with the engine producing 12 percent of its design power and each of the three refrigerators running separately at 25 percent of their design powers. All thermoacoustic phenomena were working as expected, but a crack in an inaccessible weld prevented testing at higher powers. During 2002, this system is being rebuilt, including dramatic improvements to the burner and burner-engine heat exchanger. Financial support for this effort is provided by Praxair and NETL.

# **Next Steps**

The next step in capacity will target 10,000 gpd, the largest size that we believe can be factory produced en masse and transported by rail. Initial brainstorming is underway and serious engineering design will begin soon. This effort will be financed by *Praxair* and by the Advanced Technology Program of the National Institute of Standards and Technology.

However, the development of an efficient, low-cost acoustic liquefier is challenging. Even the 500 gpd system is a scaleup of a factor of 1600 in cooling power over the first laboratory demonstration, which used simple electric heat to power the engine and an

electric-heat test load on the refrigerator, and had such poor efficiency that it would have liquefied only 9 percent of a natural-gas stream while burning the other 91 percent. Nevertheless, the 10,000 gpd system is expected to liquefy 80 percent of its throughput, and we expect that further improvements can eventually bring the efficiency close to 90 percent without compromising the low cost and reliability of the thermoacoustic approach.

#### **Back to Basics**

Readers familiar with Stirling engines or refrigerators will recognize that the processes in the regenerators and heat exchangers discussed above in the context of Figure 1 are identical to the processes in Stirling devices. Hence, another way to view thermoacoustics is as one chapter in the story of the elimination of moving parts and sliding seals from Stirling devices — a story in which earlier chapters include Beale's invention of the free-piston Stirling engine and Gifford and Longsworth's invention of the basic pulse tube refrigerator (Beale, 1969; Gifford and Longsworth, 1965). A key aspect in the thermoacoustics chapter is the deliberate use of inertial effects in the oscillating helium. A moving slug of helium can behave inertially much like a moving solid piston, bouncing against the compressibility of nearby helium to act like a spring-mounted mass. From this point of view, the half-wave resonator of Figure 1 can be thought of as if the mass of the helium in the central third of the resonator bounces resonantly against the compressibilities of the helium in the upper and lower thirds of the resonator, the resulting resonance acting like a flywheel to keep the thermoacoustic engine working from one expansion stroke to the next. The narrow portions of the system labeled

"inertance" are also local accentuators of inertial mass, enforcing the correct amplitude and time phasing of the gas motion in the nearby regenerators. Portions labeled "compliance" accentuate compressibility.

Another key aspect of the elimination of moving parts from Stirling systems is the use of pulse tubes and thermal buffer tubes in place of cryogenic or red-hot pistons. These portions of the system maintain thermally stratified adiabatic oscillating flow, thereby transmitting acoustic power from the cryogenic temperature (in a refrigerator) or the red-hot temperature (in an engine) to ambient without suffering from convective heat leak. Some current fundamental research in thermoacoustics is directed toward understanding and maintaining this thermally stratified condition in the presence of violent oscillating flow.

Efficiency and power density are two key figures of merit for any energyconversion technology. The power of thermoacoustic devices is roughly proportional to  $p_{avg}Aa(p_{osc}/p_{avg})^2$ , with  $p_{avg}$  the average pressure, A the cross sectional area of the regenerator, a the sound speed of the helium, and  $p_{osc}$  the amplitude of the oscillating pressure. Helium has the highest sound speed of the inert gases, so high-pressure helium is used in most thermoacoustic systems, including the acoustic liquefier. This leaves  $p_{osc}/p_{avg}$  as the primary variable which might be increased in order to increase power per unit area. Unfortunately, increasing  $p_{osc}/p_{avg}$ generally reduces efficiency, as a variety of higher loss processes such as turbulence grow in importance, and as the demands on heat exchangers increase. As thermoacoustics matures from scientific inquiry to realistic engineering, these are among the tradeoffs that must be made.

For more information contact the authors, Greg Swift, with Los Alamos National Laboratory at swift@lanl.gov or via telephone at 505-665-0640, or John Wollan with Praxair at John\_Wollan@praxair.com or by telephone at 303-549-7204. A more complete, animated version of Figure 1 (for PCs, not Macs) can be obtained by downloading TashOpZp.exe from http://www.lanl.gov/thermoacoustics/mov ies.html. Further background on the fundamentals of thermoacoustics is available at http://www.lanl.gov/ thermoacoustics/, which includes links to journal publications and a book. Information about the natural-gas liquefier is also available at http://www.lanl.gov/mst/ engine/econ.html.

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